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TECHNICAL REPORT ARCCB-TR-99011

LIQUID METAL EMBRITTLEMENT OF ASTM A723 GUN STEEL BY INDIUM AND GALLIUM

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JUNE 1999

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REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY *(Leave blank)*

2. REPORT DATE
June 1999

3. REPORT TYPE AND DATES COVERED
Final

4. TITLE AND SUBTITLE
LIQUID METAL EMBRITTLEMENT OF ASTM A723
GUN STEEL BY INDIUM AND GALLIUM

5. FUNDING NUMBERS
AMCMS No. 8Y10.00.0000.0

6. AUTHOR(S)
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7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
U.S. Army ARDEC
Benet Laboratories, AMSTA-AR-CCB-O
Watervliet, NY 12189-4050

8. PERFORMING ORGANIZATION REPORT NUMBER
ARCCB-TR-99011

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
U.S. Army ARDEC
Close Combat Armaments Center
Picatinny Arsenal, NJ 07806-5000

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited.

12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

An investigation was performed to determine the effects of gallium and indium on the liquid metal embrittlement of ASTM A723 gun steel. Liquid metal embrittlement tests were conducted slightly above the melting temperature of the embrittling metal in an elevated temperature furnace coupled to an Instron mechanical tester. Low cycle fatigue and monotonic tensile testing of notched tensile specimens were performed. In addition, the effect of strain rate on the embrittlement of ASTM A723 by gallium was evaluated. The low cycle fatigue tests showed a high sensitivity to liquid metal embrittlement; both the gallium and the indium significantly degraded the fatigue life of the gun steel. Monotonic tests on ASTM A723 also showed an embrittling effect with both gallium and indium, although the effect was less severe than when tested in low cycle fatigue.

14. SUBJECT TERMS
Liquid Metal Embrittlement, Environmental Cracking, Gallium
Indium, Gun Steel, High-Strength Steels, ASTM A723

15. NUMBER OF PAGES
21

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

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ACKNOWLEDGEMENT

The authors would like to thank A. Fish for her SEM and EDS analyses of liquid metal embrittled fracture surfaces.

BACKGROUND

Liquid metal embrittlement (LME) is known to cause failures in gun tubes. Circa 1977, a dramatic example of this occurred at Watervliet Arsenal during the manufacturing of a 105-mm M68 gun tube (ref 1) (Figure 1). Lead was electroplated onto the gun tube and used as a lubricant during the swage autofrettage process. During the post-autofrettage thermal treatment, the lead became molten and embrittled the gun tube, resulting in a complete transverse fracture. The embrittlement was caused by axial tensile residual stresses that were imparted during the autofrettage process. These stresses are known to be much smaller in magnitude than hoop residual stresses; this demonstrates the insidious nature of liquid metal embrittlement.

Although LME has caused many unforeseen failures, it has also been exploited as a beneficial research and engineering tool (ref 2). For example, copper and aluminum alloys have been separated into individual grains using liquid mercury and gallium, respectively. Also, American Society of Testing and Materials (ASTM) Specification B154-95 calls for LME of copper and copper alloys by mercury to detect residual stresses.

LME has also been investigated for military applications. Patent No. 4,120,701, by Walker and Steward, deals with the LME of high-strength steels using mercury amalgams. The patent states that "...another object of the present investigation is to provide a reactant...together with a projectile to defeat steel armor plate." In addition, a recent publication by the Foreign Intelligence Office (Nonlethal Technologies Worldwide, NGIC-1147-101-98) stated that LME is being assessed by other countries for potential use in nonlethal military operations.

Although the first case of LME in modern technical literature was reported in 1874 (earlier records of LME [by mercury] date back almost 2000 years [ref 3]), it remains an unfamiliar embrittlement mechanism that is not fully understood. The mechanism for LME is believed to be similar to that of hydrogen embrittlement—the liquid metal lowers the cohesive strength of the metallic bonds. As with hydrogen embrittlement and stress corrosion cracking, a combination of criteria must be met for cracking to occur. Stress concentrators such as notches increase the likelihood and severity of LME. As the temperature continues to increase above the melting point of the liquid metal, the severity of embrittlement decreases until full ductility is restored. Furthermore, although embrittlement does not typically occur much below the melting point of the embrittling species, this phenomenon—solid metal induced embrittlement (SMIE)—can occur. It has also been reported that LME is sensitive to strain rate and that higher strain rates promote a higher degree of embrittlement (ref 4). *It is essential for the liquid metal to be in intimate contact with the surface for LME to occur.* The presence of even a monotonic layer of surface oxide, for example, can preclude the occurrence of liquid metal embrittlement. When a metal does crack due to LME, the results can be quite astounding. For example, crack growth rates have been measured to be several feet per second for aluminum embrittled by a mercury amalgam (ref 5). Table 1 summarizes metals that are known to cause liquid metal embrittlement of high-strength steels.

TEST PROCEDURE

Before conducting LME tests on steels, the authors witnessed the severity of LME in known embrittlement couples. For these tests, a 2" long x 0.5" wide x 0.080" thick 2024-T3 aluminum (Al) alloy plate and pure gallium (Ga) were used. The Ga was wetted on the Al plate using only a soldering iron to melt the Ga and a metal scribe to abrade any surface oxide on the Al and facilitate the wetting. The typical amount of Ga used for these demonstrations was 30 mg; in theory, however, a thin, wetted film of a few microns thickness is sufficient to cause liquid metal embrittlement (ref 4). The plates were manually subjected to four-point bending before and after the Ga was applied. An elastic stress calculation determined that the fracture bending stress (in air) for the plate was approximately 6500 lb. Once the Ga was added, applied loads of only a few pounds were needed to completely fracture the Al. Rather than cracking through the plate, the cracks ran the length of the plate, indicating the strong texture that is common to aluminum plate. In fact, in further tests the aluminum cracked without any load being applied. Grain boundary diffusion and embrittlement (ref 5) is a unique LME mechanism that is active in the Al-Ga couple.

All subsequent LME tests were conducted on ASTM A723 steel. Due to availability, cost and time restraints, LME tests with Ga were conducted on ASTM A723 Grade 2 steel (120-mm M256 material), and LME tests with indium (In) were conducted on ASTM A723 Grade 1 steel (155-mm M284 material). Both types of ASTM A723 steel were re-heat treated in small sections to a yield strength of approximately 190 Ksi. Table 2 contains the typical chemistry of the tested ASTM A723 steels.

Liquid metal embrittlement tests were conducted near the melting temperature of the embrittling metal in an elevated temperature furnace coupled to an Instron mechanical tester. Both low cycle fatigue and monotonic tensile testing of notched tensile specimens were performed (Figure 2).

Ga was successfully wetted onto the steel surface by first electropolishing the test specimen in a mixture of ethyl alcohol (23.5% by volume) and phosphoric acid (66.5% by volume) for approximately 1-2 minutes at a current density of approximately 300 ma/in^2. The In was successfully wetted onto the steel surface by applying a ZnCl flux agent to the notch root, heating the specimen to approximately 375°F, wrapping the notch in the specimen with 0.005 in. thick In foil, and using a soldering iron to melt the In and break any surface oxide present.

A low cycle fatigue life of approximately 5000 cycles was initially established to represent the stresses and strains a gun tube may be subjected to during its service life. The Neuber notch method was employed to calculate the load range for specimens to fail by fatigue in laboratory air at 5000 cycles (ref 8). As a result of that analysis, it was determined that a maximum load (P_{max}) of 5500 lb. should be used. The ratio of minimum to maximum load (R ratio) was 0.1, and the test frequency was 5 Hz. Monotonic tests were conducted at stroke rates of 0.003 in./min. and 0.3 in./min. for the Ga on ASTM A723 and 0.003 in./min. for the In on ASTM A723. All tests on the Ga and In were conducted at temperatures of 95 and 325°F, respectively. A data acquisition system was used to record load versus time for all monotonic tensile tests on ASTM A723.

RESULTS

The cyclic tests conducted on ASTM A723 steel and Ga are summarized in Table 3. Note that the average cycles to failure for the ASTM A723 steel were 5682, approximately 12% higher than the 5000-cycle prediction based on the Neubers notch method. The average cyclic life of the embrittled steel by Ga was 1323 cycles, or a reduction in fatigue life of 77%. On the fracture surfaces of the embrittled steel specimens, there was obvious wetting of the Ga; in fact, the Ga could not be removed easily by ultrasonic cleaning, acetate tape, or electropolishing. It was, therefore, very difficult to examine the underlying fracture surface under the scanning electron microscope (SEM). However, there was evidence of intergranular fracture— a brittle fracture mode in steel (Figure 3). A corresponding energy dispersive x-ray analysis (EDX) revealed a high concentration of Ga (Figure 4). The fracture mode was ductile (microvoid coalescence) and dramatically different in appearance (Figure 5) in an area of the specimen that was not embrittled. The predominant fracture mode for this gun steel tested under "normal" conditions is ductile, i.e., microvoid coalescence. The EDX from the ductile fracture region showed no presence of Ga (Figure 6).

The cyclic tests conducted on ASTM A723 steel and In showed that the liquid metal severely affected the low cycle fatigue life of the steel. In all cases, cracking occurred before the load reached P_{max} of 5500 lb. As with ASTM A723 specimens tested with Ga, there was evidence of intergranular cracking where the In was present and microvoid coalescence when it was not (Figures 7 and 8).

The monotonic tensile tests conducted on ASTM A723 and Ga at a stroke rate of 0.003 in./min. exhibited a lower load to failure. Again, there was evidence of wetting of Ga onto the fracture surface. The tests conducted at 0.3 in./min. did not indicate any embrittlement even though wetting was evident on the fracture surface. These results are contrary to some literature that states that the higher the stroke/strain rate, the higher the degree of embrittlement (ref 4). Figures 9 and 10 show the effects of slow and fast strain rate tests on ASTM A723 with Ga. The lower load to failure described by the Embrittlement Index (EI) is $1-(load_{w/environment}/load_{w/o\ environment})$. Therefore, the closer the EI is to unity, the greater the degree of embrittlement. The EI for the slow strain rate test was 0.34 and zero for the fast strain rate test of Ga and ASTM A723.

The monotonic tests conducted on ASTM A723 and In at a stroke rate of 0.003 in./min. also resulted in embrittlement. The EI of 0.52 illustrates that ASTM A723 is more susceptible to liquid In than liquid Ga (Figure 11).

DISCUSSION

Although the strength of the ASTM A723 Grade 1 and Grade 2 material was equal, the difference in chemistry and processing could have biased the test outcome. ASTM A723 Grade 1 steel (155-mm M284 material) is electric furnace melted and vacuum degassed (EFM-VD); ASTM A723 Grade 2 steel (120-mm M256 material) is electric furnace melted and electroslag remelted (EFM-ESR). The M284 material is more heterogeneous than the M256; contains a high level of impurities; and has lower reduction in area (%RA), Charpy impact energy (CVN), and fracture toughness. These characteristics may result in a higher susceptibility to environmental cracking. Previous hydrogen cracking tests (Vigilante, et al. [ref 9]) demonstrated that when 155-mm M284 material was tested at the same strength level and test conditions as 120-mm M256, the incubation time was noticeably shorter for M284—one measure of susceptibility to hydrogen cracking.

SUMMARY

- Liquid metal embrittlement (LME) has occurred in gun tubes. In 1977, a gun tube fractured completely due to liquid lead and residual tensile stress from the swage autofrettage process.

- Under specific conditions, LME can be an insidious and catastrophic failure mechanism by which cracks can grow on the order of meters per second.

- In order for LME to occur, intimate contact between a specific embrittling metal and a specific host metal must occur (wetting). In addition, as with all cracking, a tensile stress—either applied or residual—must be present. When LME does occur, the most severe embrittlement occurs near the melting temperature of the liquid metal.

- The effects of liquid gallium (Ga) and liquid indium (In) on the embrittlement behavior of ASTM A723 steel were investigated. Notched tensile bars were tested both monotonically and cyclically at slightly above the melting temperature of Ga and In.

- The cyclic tests on the ASTM A723 specimens tested with both Ga and In exhibited a greater sensitivity to liquid metal embrittlement than the monotonic tests.

- In cyclic tests of Ga on ASTM A723, the fatigue life was degraded by 77%. In slow strain rate monotonic tests (0.003/min.), the load to failure was reduced by 34% (EI = 0.34). No reduction in load was observed during the fast strain rate (0.3/min.).

- In cyclic tests of In on ASTM A723, the fatigue life was essentially nil because the maximum load amplitude was never reached during testing. In slow strain rate monotonic tests, the load to failure was reduced by 52% (EI = 0.52).

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Table 1. Melting Temperatures of Various Liquid Metals Known to Embrittle High-Strength Steels (ref 2)

Liquid Metal	Melting Temperature (°F)
Mercury (Hg)	-38
Gallium (Ga)	85
Indium (In)	313
Lithium (Li)	356
Tin (Sn)	449
Cadmium (Cd)	610
Lead (Pb)	620
Zinc (Zn)	787
Tellurium (Te)	841
Antimony (Sb)	1167
Copper (Cu)	1981

Table 2. Typical Chemistry of ASTM A723 Steels Tested (wt. %)

	C	Ni	Cr	Mo	V	Mn	S	P
A723 Grade 1 (155-mm M284)	0.32 - 0.36	2.1 - 2.25	0.9 - 1.1	0.45 - 0.55	0.09 - 0.12	0.55 - 0.65	0.008 max.	0.010 max.
A723 Grade 2 (120-mm M256)	0.32 - 0.36	2.5 - 3.2	0.9 - 1.1	0.45 - 0.55	0.09 - 0.12	0.55 - 0.65	0.004 max.	0.008 max.

Table 3. Summary of Cyclic LME Test Results

Environment	Test Temperature (°F)	Fatigue Life, N_f (no. of cycles to failure)
None	98	6683
None	98	4957
None	98	5564
None	98	5525
Ga	98	1347
Ga	98	1299
None	320	4432
None	320	4641
In	320	0.25*
In	320	0.25*

**The specimens failed at a load amplitude of approximately 4200 lb., before the peak tensile load amplitude was reached.*

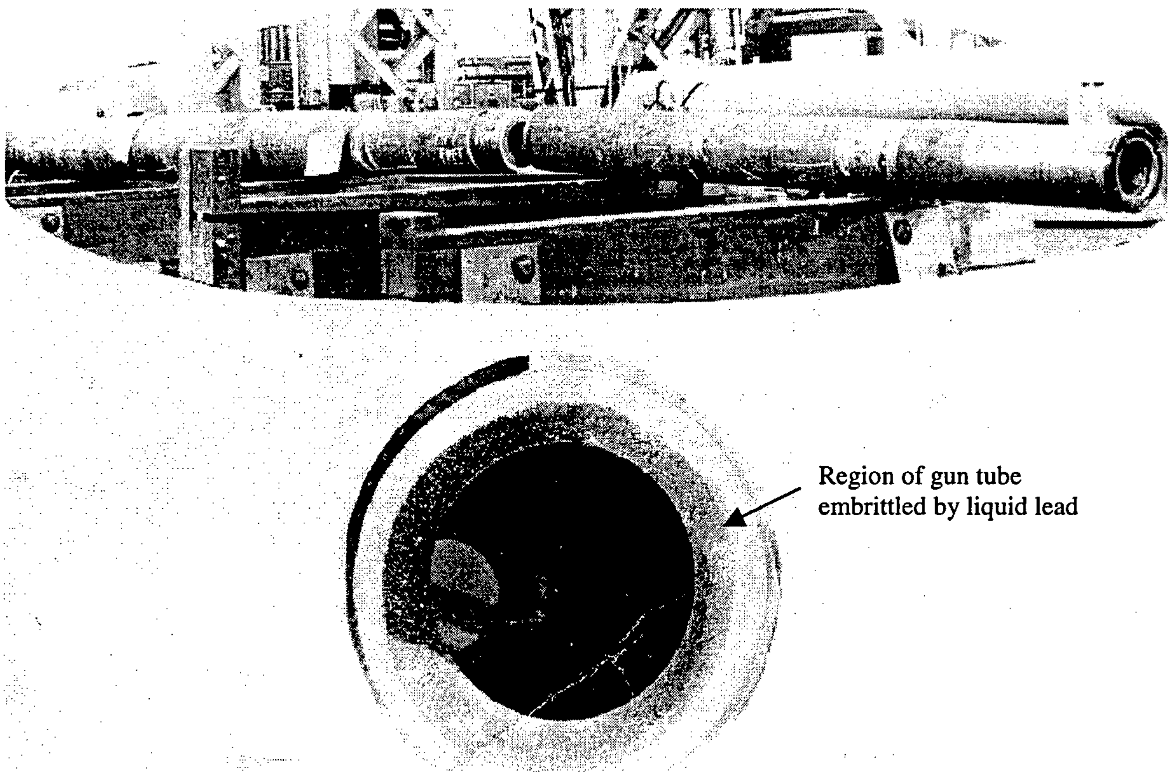

Figure 1. Liquid metal embrittlement failure of a 105-mm M68 gun tube due to lead.

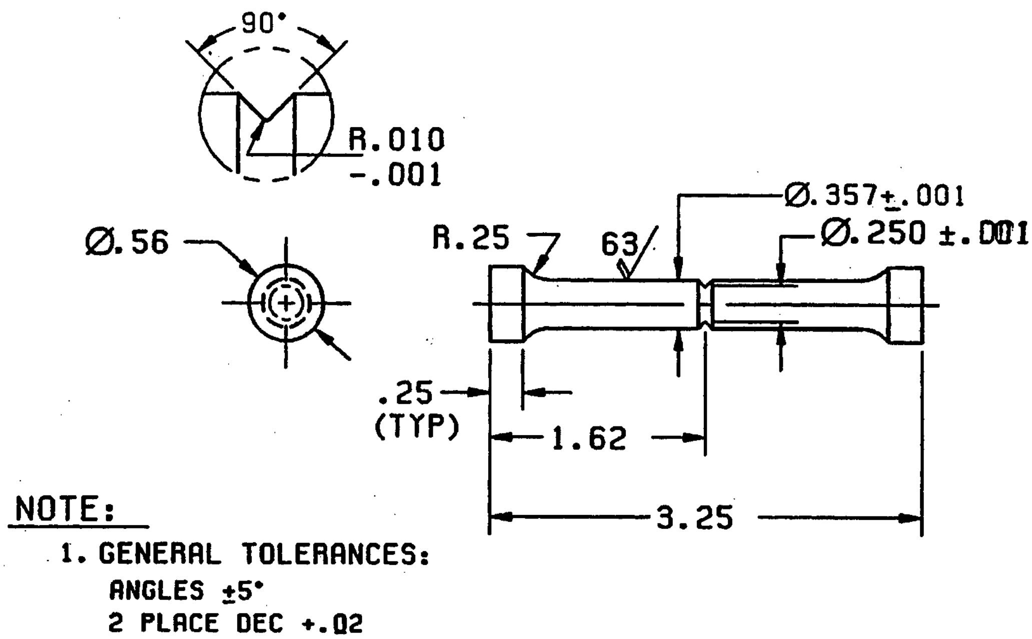

Figure 2. Mechanical drawing of a notched tensile specimen.

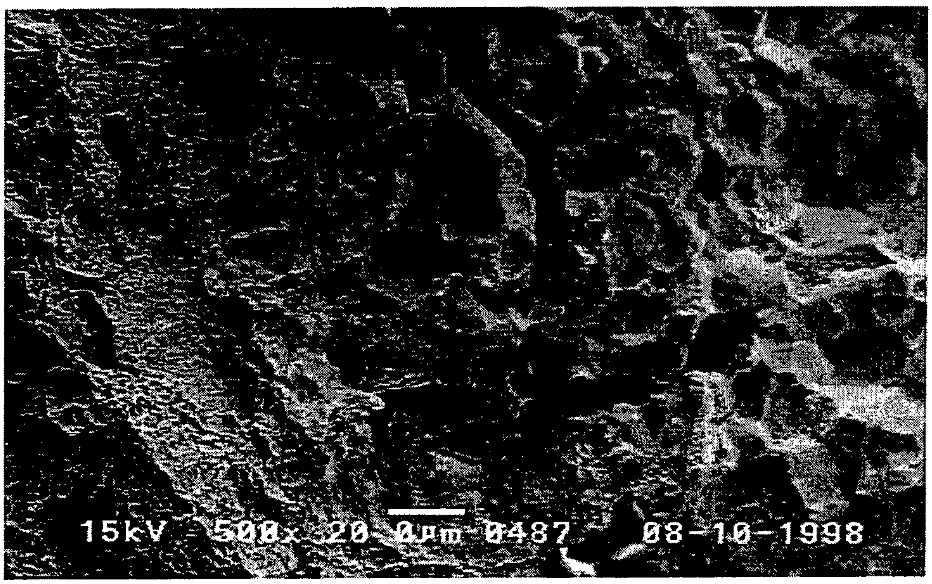

Figure 3. Brittle intergranular fracture surface (right) evident in the LME of ASTM A723 by Ga (mag 500x).

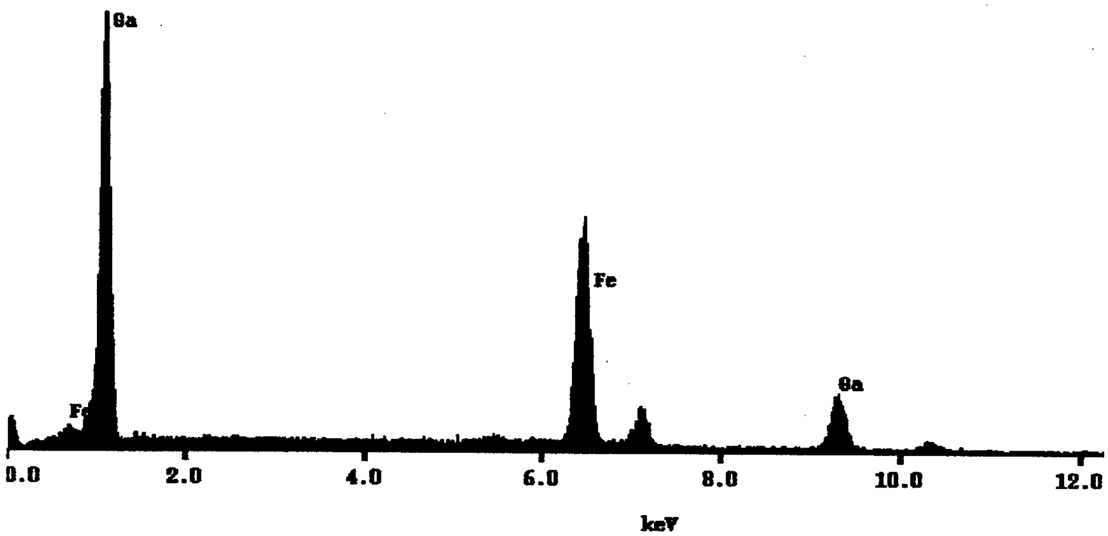

Figure 4. EDS of a region of the ASTM A723 fracture surface embrittled by Ga.

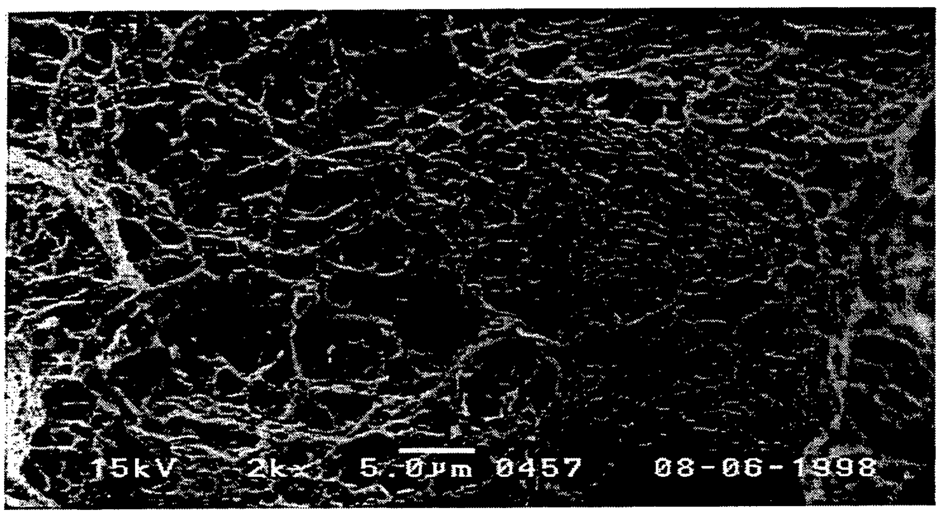

Figure 5. Ductile microvoid coalescence fracture surface—the normal fracture surface appearance in ASTM A723 (mag 2000x).

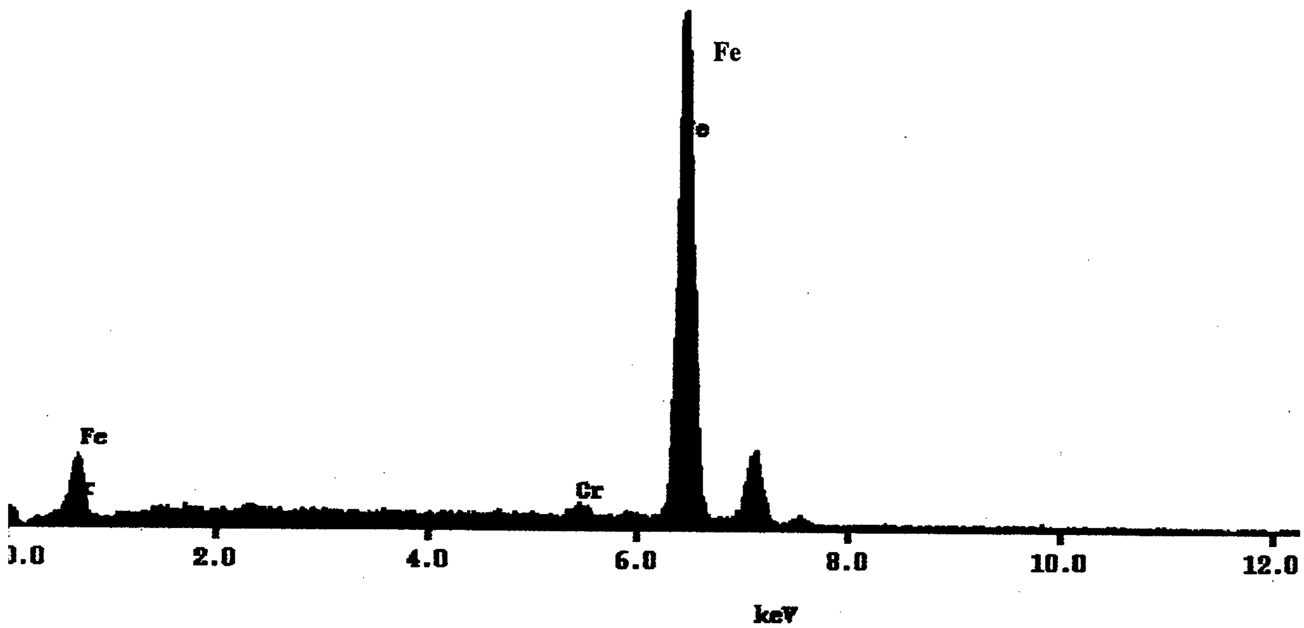

Figure 6. EDS of a region of the ASTM A723 fracture surface not embrittled by Ga.

Figure 7. Brittle intergranular fracture of ASTM A723 due to LME from In (mag 700x).

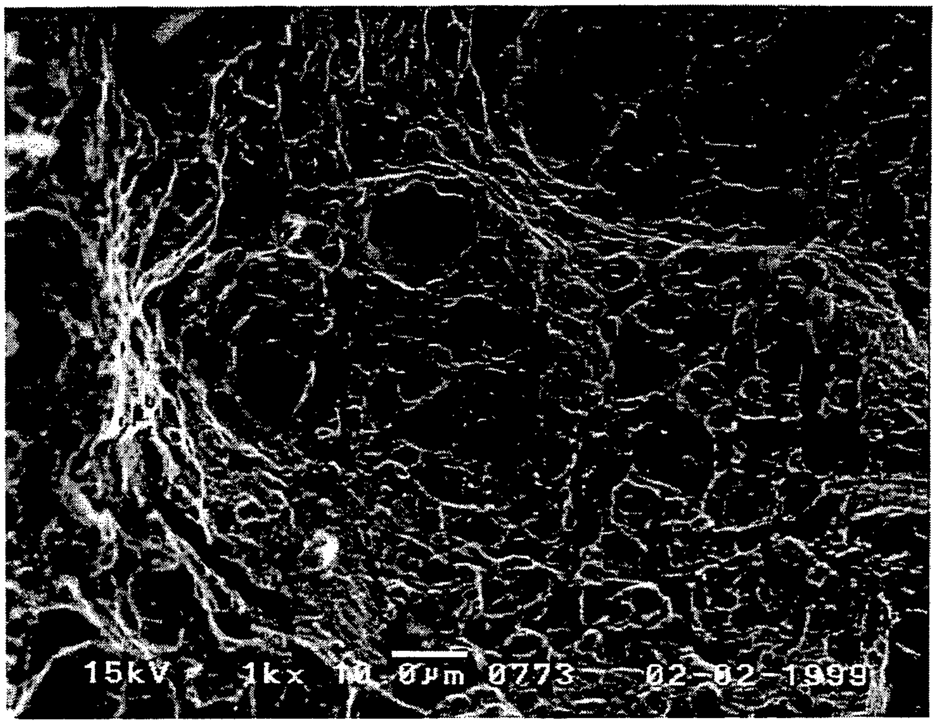

Figure 8. Microvoid coalescence in region of ASTM A723 fracture surface not wetted by In (mag 1000x).

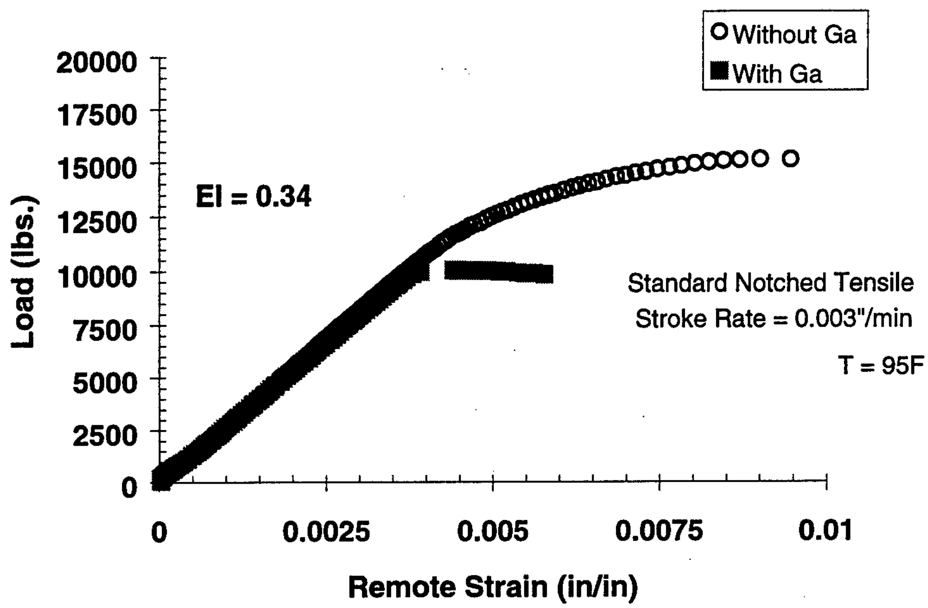

Figure 9. Effect of liquid Ga on the monotonic slow strain rate tensile behavior of ASTM A723 steel.

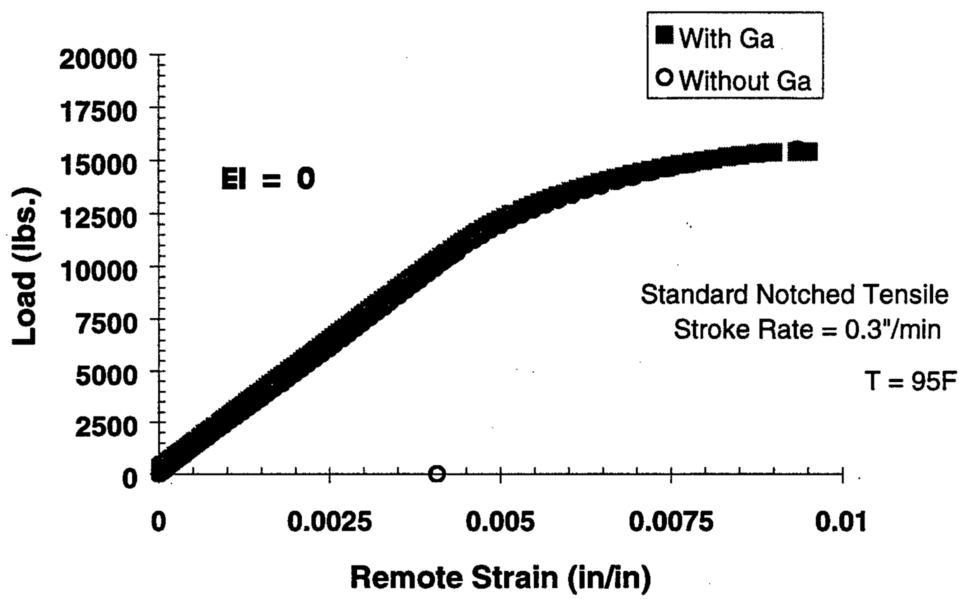

Figure 10. Effect of liquid Ga on the monotonic high strain rate tensile behavior of ASTM A723 steel.

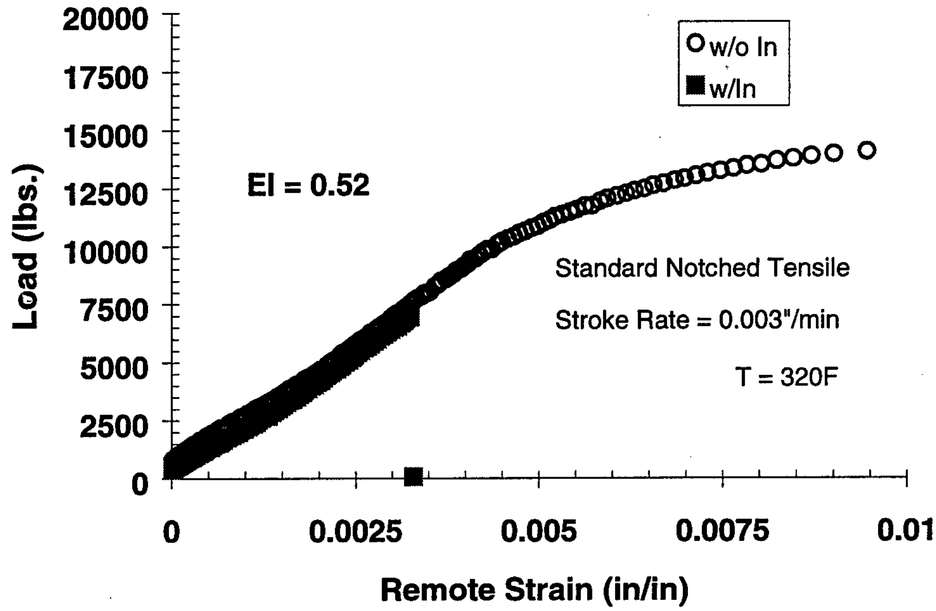

Figure 11. Effect of liquid In on the monotonic slow strain rate tensile behavior of ASTM A723 steel.

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